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Architectures for a Spin Quantum Computer Based on Endohedral Fullerenes

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We present a discussion of recent concepts for the construction of a spin quantum computer using endohedral fullerenes. The fullerene molecule is a static, room-temperature trap for atoms with slowly relaxing electron and nuclear spins. The fullerene "containers" can be used to arrange the spins in complex structures such as a linear chain, to form a spin quantum *register*. We discuss the probable properties of such registers and different strategies to use them in a quantum computer design, including gating and read-out methods.

1. Introduction Quantum computation using nuclear spins has been demonstrated in a variety of experiments [1, 2] but is believed to be limited to a small number of qubits [3]. Proposals for realistic spin quantum computer architectures have to address the question of scalability [4]. There are two widely cited concepts fulfilling this criterion, by Kane [5] and by Loss and DiVincenzo [6]. Both concepts are based on electric-field controlled exchange interaction between electron spins, which might however be very difficult [7].

Recently, Harneit [8], Suter and Lim [9], and Twamley [10] presented concepts for quantum computation using endohedral fullerenes as spin-qubits, and a microwave-pulse controlled magnetic dipolar interaction between qubits. A symbolic drawing synthesizing these concepts is shown in Fig. 1. In this paper, we present the current knowledge about the qubits in question, and we discuss the proposed operational schemes and computer architectures that might be scalable.

2. Endohedral Fullerenes as Qubits

2.1 The spin system of an endohedral fullerene Free nitrogen and phosphorus atoms possess a half-filled p-shell and thus a total electron spin $S = 3/2$. They can be encapsulated in the cage-like fullerene molecules C_{60} and C_{70} whilst retaining their atomic character [11]. At strong magnetic fields, the four possible values $m_S = -3/2, -1/2, +1/2$, and $+3/2$ for the electron spin z-component give rise to four equidistant energy levels. The total spin system also contains the nuclear spin, viz. $I = 1/2$ for ^{15}N and ^{31}P, and $I = 1$ for the abundant nitrogen isotope ^{14}N. The hyperfine interaction A between electron and nuclear spin splits every electron spin level into two (^{14}N: three) sub-levels. The energy level diagram and electron spin resonance patterns are shown in Fig. 2.

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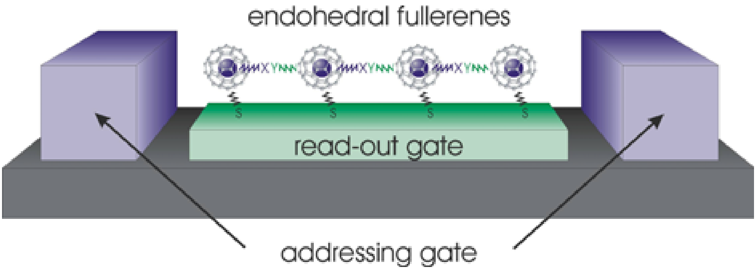

Fig. 1 (online colour). Scheme for a solid-state spin quantum computer based on linear chains of endohedral fullerenes. The qubit is encoded in the electron–nuclear spin system of a paramagnetic atom trapped in a fullerene (this complex is called an "endohedral fullerene"). Qubit coupling is achieved by magnetic dipolar interaction between adjacent endohedral electron spins. Universal quantum gates are realized by magnetic resonance pulses. A pair of micron-sized wires produces a magnetic field gradient along the linear fullerene chain for local addressing. SWAP operations between nuclear and electron spins could effectively turn on and off next-neighbor interactions. The single-spin read-out gate is symbolic and needs to be developed. The quantum register part can also be used for a quantum cellular automaton if many identical fullerene chains are built whose endohedral atoms follow a certain pattern (ABAB... or ABCABC...)

The six (^{14}N: nine) allowed ESR transitions fall on two (^{14}N: three) lines since the electron spin levels belonging to the same nuclear spin value are equidistant, indicating that there is no distortion or transfer of the electron spin density form the encapsulated atom to the fullerene cage. The ESR lines are very sharp even for a powder sample containing some (10^{-4}) filled cages in a polycrystalline matrix of empty fullerenes, again showing that the spin system is highly shielded from the environment.

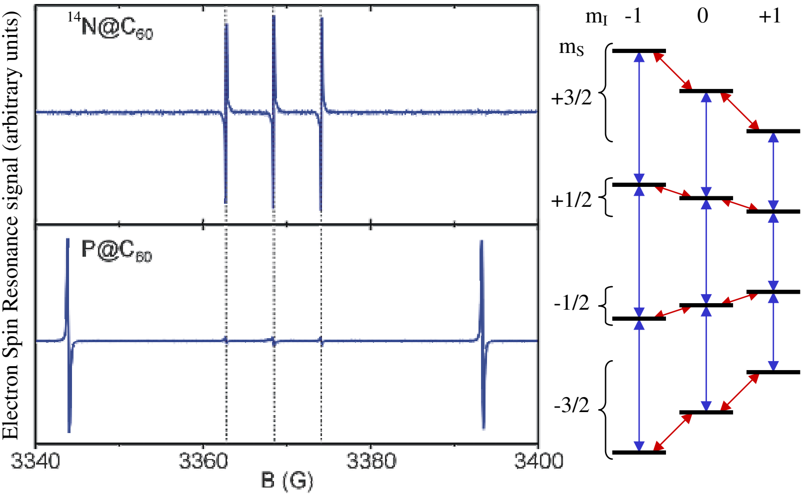

Fig. 2 (online colour). Spin system of the group-V endohedral fullerenes. Left: electron spin resonance patterns for dilute ^{14}N@C$_{60}$ and ^{31}P@C$_{60}$ powders at room temperature [12]. Right: energy level scheme of the coupled electron–nuclear spin system for ^{14}N@C$_{60}$ (not to scale). The vertical arrows indicate allowed ESR transitions for which m_I = const and $\delta m_S = \pm 1$. The horizontal arrows show allowed NMR transitions for which m_S = const and $\delta m_I = \pm 1$. A similar scheme holds for ^{31}P@C$_{60}$ and ^{15}N@C$_{60}$. These atoms have a nuclear spin $I = 1/2$, and thus only two sub-levels $m_I = \pm 1/2$ for each electron spin level

Table 1
Spin properties of the endohedral C_{60} fullerenes with electron spin $S = 3/2$ [14–16]

endohedral fullerene	nuclear spin I	hyperfine constant A (MHz)	electron spin relaxation in dilute powders			
			$T = 300$ K		$T = 5$ K	
			T_1 (μs)	T_2 (μs)	T_1 (s)	T_2 (μs)
^{14}N@C$_{60}$	1	15.88	120	20	0.5–9[a])	20
^{15}N@C$_{60}$	1/2	22.26	45	11	1	14
^{31}P@C$_{60}$	1/2	138.4	2.7	1.3	0.5–8[a])	14–28[b])

[a]) depending on sample quality
[b]) depending on endohedral fullerene concentration

The spin Hamiltonian H for a single endohedral fullerene with electron spin $\mathbf{S}$ and nuclear spin $\mathbf{I}$ in a large magnetic field $\mathbf{B}$ is given by (we set $\hbar = 1$)

$$H = \mathbf{B}(\gamma_n \mathbf{I} - \gamma_e \mathbf{S}) + \mathbf{I}A\mathbf{S} + \mathbf{S}D\mathbf{S} \,. \tag{1}$$

The nuclear Zeeman $\gamma_n \mathbf{BI}$ term is always small compared to the electron Zeeman term $\gamma_e \mathbf{BS}$ and will be dropped henceforth, since the gyromagnetic ratio γ_e of the electron is much larger than that of the nucleus, γ_n. The hyperfine term $\mathbf{I}A\mathbf{S}$ couples the nuclear spins to the electron spins, A is a number characteristic of the endohedral atomic species (see Table 1). The fine structure or zero-field splitting term $\mathbf{S}D\mathbf{S}$ is only important when the fullerene cage is not a sphere, i.e., $D = 0$ for undistorted C_{60} fullerenes and even for C_{70} molecules rapidly tumbling in a liquid. In a $(C_{60})_2$ dimer (a.k.a. C_{120}), an atom enclosed in one of the bound C_{60} cages finds a highly non-spherical environment and the zero-field splitting term can be as large as 15 MHz [13].

2.2 Relaxation times The electron spin of endohedral fullerenes has a remarkably long lifetime that makes it a useful embodiment of a qubit for quantum computation. The temperature dependent spin–lattice relaxation time T_1 and spin–spin relaxation time T_2 of the electron spin have been measured for magnetically dilute powder samples of N@C$_{60}$ [14, 15] (see Fig. 3) and P@C$_{60}$ [16]. While T_2 is practically independent of temperature and about 20 μs for these molecules, T_1 increases with decreasing temperature from about 100 μs at $T = 300$ K to several seconds below $T = 5$ K. The relaxation times are of the same order for solid and solution samples at room temperature (see Table 1).

The main spin-lattice relaxation path is due to the vibration of the enclosed atom in the fullerene shell and thus quite independent of variations of the exterior world. The spin–spin relaxation mechanism is probably foremost due to incoherent dipolar interaction with electron spins of other endohedral fullerenes. In a careful quantum computer design this interaction should be controlled and thus the effective decoherence time could be much longer than the apparent T_2.

The nuclear spin system has been studied in much less detail. The main reason is the difficult synthesis of the molecules that has concentrated on ^{14}N@C$_{60}$ so far, a nuclear species with low NMR sensitivity. As a consequence, the nuclear spin relaxation times are at present unknown.

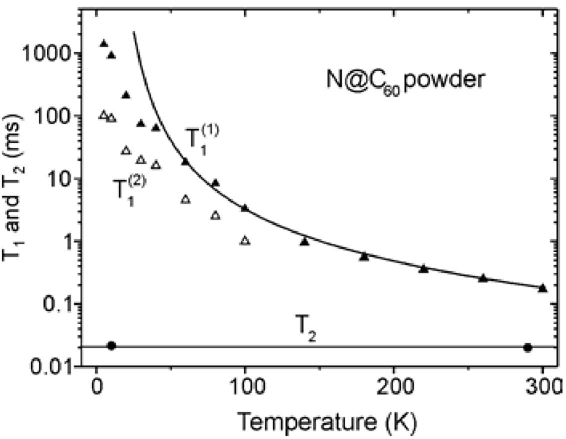

Fig. 3. Relaxation times T_1 and T_2 of N@C_{60} powder, after [15]. For the spin–lattice relaxation time, a bi-exponential decay is observed ($T_1^{(1)}$, $T_1^{(2)}$). The solid line for T_1 is a model calculation assuming only modulation of the hyperfine constant corresponding to an endohedral nitrogen atom rattling around in the fullerene cage with an oscillator energy of 13 meV. The temperature independent coherence time T_2 is at present unexplained and might be due to unresolved spin–spin interactions between endohedral electron spins (presumably of dipolar origin). If that is the case, the actual T_2 relevant for quantum computation might be longer for a careful arrangement of endohedral fullerenes in a quantum register

2.3 Spin–spin coupling Experimental studies on the concentration dependent line broadening [17] as well as theoretical calculations [18] show that the exchange interaction between two endohedrals, either directly by contact or mediated by a substrate is vanishing small, thus leaving the magnetic dipole interaction as the only strong coupling between two endohedrals. For two adjacent endohedrals at a center-to-center distance of 1 nm (touching fullerene shells) the magnetic dipole interaction between the electron spins can be as large as 52 MHz.

2.4 Coupling to external fields The coupling of the spin system to external fields is important because it opens the path to coupling control and read-out strategies. Indeed, one of the main attractions of Kane's concept for a spin quantum computer [5] is the vision of entirely local addressing and read-out using only electric fields. The general desideratum is to have an external field (i.e., a classical variable) that tunes some parameter of the spin Hamiltonian, e.g., the qubit transition energy (A-gate), the qubit–qubit coupling, (J-gate), or the qubit–detector coupling (read-out). On the other hand, the coupling to the external field should not be so strong as to perturb the fragile superposition states of the quantum system, and the spatial extension of the gating fields has to be carefully controlled.

We have investigated the effect of an external electric field on the endohedral qubits. The experiments indicate that there is no quadrupole moment for the nuclear spin [19], and no induced hyperfine-shift. These results are corroborated by simulations [20] that show that any electric field will be completely shielded by the electron system of the fullerene cage.

The only "tunable" effect so far is the fine structure splitting of the ESR lines that is seen for distorted fullerene cages [11]. One might be able to manipulate this splitting with an applied electric field, if a permanent electric dipole is attached to the fullerene molecule in such a way that it orients the symmetry axis. Another way is to use a liquid crystal matrix to the same effect. In a recent liquid crystal study [21], we were able to induce a D-splitting term in N@C_{60} and N@C_{70} molecules.

3. Computer Architectures

3.1 Molecular spin quantum register As a general strategy to actually build a quantum register we propose here to use a macro-molecular chemical approach rather than physical methods. The main reason is the relatively low thermal stability of the endohedral complexes [22]. Above 50–100 °C, the enclosed atom starts to form chemical bonds to the inside of the fullerene cage. This is well below the sublimation temperature of fullerenes so that it is at present impossible to get the endohedral fullerenes into the vapor phase, the starting point of all physical micro-structuring methods.

Fullerene chemistry, on the other hand, has developed a number of ways to engineer molecular assemblies containing fullerenes. Our endohedral fullerenes are stable under chemical reactions [23], provided they do not involve high temperatures. It is therefore attractive to combine endohedral fullerenes and fullerene chemistry to produce a spin quantum register [8]. The register may be a linear chain or even more complex. The chain may contain different endohedral species A, B, C (e.g., ^{14}N@C_{60}, ^{15}N@C_{60}, ^{31}P@C_{60}) in a sequence controlled by chemical engineering, see the article by Meyer et al. in this volume [24].

The general Hamiltonian for our spin quantum register (a linear chain of N evenly spaced endohedral fullerenes) is

$$H = \sum_{k=1}^{N} \left(\Omega_k + \sum_{i=1}^{N} \mathbf{S}_i J_{ik} \right) \mathbf{S}_k \tag{2}$$

with an addressing part $\Omega\mathbf{S}$ and a coupling part $\mathbf{S}J\mathbf{S}$. Using several endohedral species with different hyperfine coupling A_k, the addressing operator is

$$\Omega_k = -\gamma_\mathrm{e}\mathbf{B}_0 + A_k\mathbf{I}_k \tag{3}$$

and the coupling operator is

$$J_{ik} = \frac{1}{2}\, J_0(3\cos^2\vartheta - 1) \times \left(\delta_{ik}\, \frac{D_0}{J_0} + (1 - \delta_{ik})\, \frac{1}{|i-k|^3} \right) . \tag{4}$$

The dipolar coupling strength depends on the lattice spacing r_0 as $J_0 = \hbar\gamma_\mathrm{e}/r_0^3$ (D_0 will be discussed below). The dipolar coupling is not confined to nearest neighbors but rather scales as the inverse of the third power of the distance between the qubits under study. This problem has been investigated by Goldman et al. [25]. Experimentally, refocusing schemes known from NMR can be used to average out higher-order terms so that in practice, one can consider only next neighbors [9].

Note that in general, there is a self-coupling term D which has the same functional dependence on the angle ϑ between the line connecting two fullerenes and the external magnetic field as the dipolar two-qubit coupling J. By adapting the geometrical arrangement, the coupling strengths J_0 and D_0 can be designed within the following limits: The maximum coupling strength at a minimum inter-fullerene distance of 1 nm, and at an angle of $\vartheta = 0$, is $J_0 = 52$ MHz. Using special arrangements and/or chemical adducts, the self-coupling can be made as large as $D_0 = 15$ MHz (as in the case of a C_{120} dimer [13]) or kept at a value close to zero (as in the case of a symmetrical hexa-adduct [23]).

3.2 *Gated quantum register operation* For the sake of clarity, in this section we consider only the case of vanishing self-coupling, $D_0 = 0$, and regard the electron spin as an $S = 1/2$ system. These simplifications will be discussed separately in Section 3.3.

3.2.1 Addressing individual electron spins A molecular quantum register consisting of *identical* endohedral fullerenes can be locally addressed if one applies a magnetic field gradient along the spin chain [9]. This can be achieved by placing the chain perpendicularly between two wires (see Fig. 4). A current I flowing in the same direction through the wires placed at $\xi = -1$ and $\xi = +1$ produces a magnetic gradient field $B = B_g \xi(1 - \xi^2)$, where $B_g = -(\mu_0/4\pi)\, 2I/L$, and L is the distance between the wires. The new addressing operator for the electron spins then reads

$$\Omega_k = -\gamma_e \left(B_0 + B_g \frac{\xi_k}{1 - \xi_k^2} \right) + A\mathbf{I}_k\,, \qquad 0 < \xi_k < 1\,. \tag{3a}$$

The difference in magnetic field between neighboring electron spins produces a spectroscopic splitting that can even be made larger than the hyperfine splitting of an individual endohedral fullerene, so that the last term in Eq. (3a) can be dropped. The resonance frequency of a qubit (its 'address') is then given by its position ξ_k in the chain alone.

A benefit of a spin resonance frequency varying along the chain is that spin diffusion is suppressed, i.e., one can neglect the flip–flop terms in the coupling Hamiltonian $(S_{1+}S_{2-} + S_{1-}\, S_{2+})$ and truncate the dipolar interaction term to $J(\vartheta)\, S_{1z}S_{2z}$. Then, one can implement a controlled-NOT (CNOT) and any other quantum operation between adjacent electron spins using ESR analogues of well known NMR pulse sequences [1].

3.2.2 Computing with both, nuclear and electron spins Although the addressing scheme suffices in principle to make our quantum register a (electron spin) computer, the number of gate operations might be enhanced if one uses the nuclear spins as a quantum *memory* with longer coherence time and the electron spins as a *working* register only [9]. The typical length of an electron spin CNOT has been estimated [8] to be 50–100 ns, leaving time for about 200–400 of such operations within the electron dephasing time

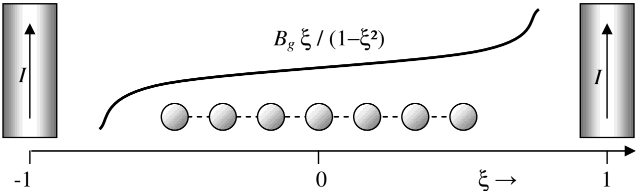

Fig. 4. Arrangement for local addressing of endohedral fullerenes in a linear chain, after [9]. A current I flowing in the same direction through two wires placed at $\xi = -1$ and $\xi = 1$ produces a magnetic gradient field $B = B_g\xi/(1 - \xi^2)$, where $B_g = -(\mu_0/4\pi)\, 2I/L$ (L is the real-world distance between the wires)

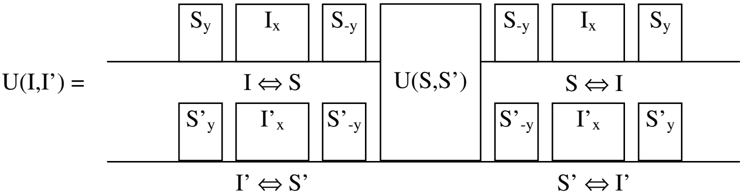

Fig. 5. A unitary two-qubit operation U on the nuclear spins **I**, **I**′ can be performed by acting on the electron spins **S**, **S**′ if one places this operation between appropriate SWAP operations **I** ⇔ **S** etc. The SWAP operations consist of three transition selective π-pulses around the indicated axes. In the figure they are chosen in a symmetrical form that automatically undoes any unwanted evolution applied to nearby 'passive' qubits [9]. The length of the π-pulses rotating the nuclear spins is much longer (some μs) than all operations performed on the electron spins, and could limit the usefulness of this scheme. The total sequence, however, is much faster than direct manipulation of the nuclei using qubit-selective pulses which would take some milliseconds

$T_2 \sim 20$ μs. Another advantage of harnessing the nuclear spins for quantum computation is that one can effectively 'switch off' (i.e. decouple) the dipolar interaction between electron spins as long as the quantum information is stored in the nuclear spins.

The main new feature needed is a SWAP operation between electron and nuclear spin. This can be brought about by using a sequence of three transition selective π-pulses that uniquely address a single electron spin. A possible scheme to implement any unitary transformation for the logical (nuclear) qubits by using their attached working (electron) qubits is shown in Fig. 5.

3.2.3 Read-out A direct read-out method of the spin information encoded in a single endohedral fullerene is still lacking. Different methods have been proposed in the literature to read single spins [9], but few experimental results have been presented so far (e.g., [26]). The main idea is to convert the spin information to some other, higher energy system, e.g., a charge in a single electron transistor, a photon in an ODMR experiment, or even just a larger spin system like a magnetic particle. The propagation of spin information along a chain using successive SWAP operations might be useful for this purpose since it permits to decouple a delicate quantum computing register from a part that is read out using methods that are likely to induce decoherence.

At present, the only read-out method available is electron spin resonance itself, which works fine but is limited to ensembles containing a large number of identical quantum registers working in parallel, just like in the case of liquid NMR computing. The situation is somewhat better in our case since we are dealing with electron spins instead of nuclear spins so that the thermal populations are larger and the ensemble can be made smaller. However, the limitation is exponential in the register size so that a single spin read-out remains an important field of study for spin quantum computing. Nevertheless, until the advent of single-spin read-out the quantum register can be tested with an ensemble type read-out.

3.3 Quantum cellular automaton operation The molecular quantum register can be built to conform to the requirements of Quantum Cellular Automaton (QCA) operation [27, 28], i.e., it can be made of spin pairs AB–AB-... or triplets ABC–ABC-... The QCA computational model has the attraction that it requires neither local addressing nor interaction switching. On the other hand, QCA computing with AB-chains only is costly in terms of resources (i.e., number of spins). For the case of ABC-chains [27], each spin corresponds to one qubit, whereas for the AB-chains [28] four spins encode one qubit. The sequential synthesis route proposed here could in principle lead to a large number of the required long spin chains, making ensemble-type read-out feasible.

Possible QCA pulse sequences for endohedral-fullerene quantum registers have been investigated by Twamley [10] who also studied the special issues arising from the fact that we have a spin-3/2 chain. An analysis in terms of an 'inner' ($|m_S| = 1/2$) and an 'outer' ($|m_S| = 3/2$) qubit proves that it is very useful to have the additional nuclear spin degrees of freedom for universal QCA quantum computation. In the absence of a spectroscopic splitting of the spin-3/2 sub-levels (i.e., when there is no fine-structure, $D_0 = 0$) it even turns out to be necessary to have access to a spin-1/2 nucleus in order to perform all single-qubit rotations. As a result, QCA quantum computing with endohedral fullerenes as suggested in [10] requires using the nuclear spins as the qubits and the electron spins as a 'bus qubit'.

An estimate of the *logical* gate duration of ~30 μs was given in [10], which might just be possible within the presently known decoherence time of the electron spins. As stated in Section 2, we know little about the nuclear dephasing times and whether the measured T_2 of the electron spins represents the practical limit for quantum computation. The effective dephasing times in a quantum register molecule with a well-defined coupling might be much longer and could be assessed by measuring endohedral fullerene dimers in an orienting liquid-crystalline matrix.

4. Conclusion Endohedral fullerenes have promising properties for the experimental realization of a spin quantum computer. Even at room temperature, some hundreds of basic logic gates might be possible using the electron spins as the qubits and the nuclear spins for addressing purposes. The nuclear spins of the endohedral atoms probably have longer relaxation times than the electron spins so that more operations could be performed using the nuclear spins as a 'qubit memory' and the electron spins as 'bus qubits'.

A spin quantum register could be built by chemical means using the cage-like fullerene molecules as a versatile 'handle' to the atom-like spin system of the encapsulated nitrogen or phosphorus. The register molecule can be tested first in suitable liquid-crystalline matrices using ensemble-type measurements that have proven so useful in liquid-NMR quantum computation. When single-spin read-out methods compatible with endohedral fullerenes are found the same register molecule can be placed on a substrate and local gates can be implemented.

We have presented a quantum register operation model for such a solid-state implementation that is somewhat reminiscent of Kane's quantum computer [5]. The main difference is that our spin system is embedded in a molecular rather than in a bulk substrate. This choice actually prevents the use of local electric fields for gating, since the fullerene molecules act as a Faraday cage. We have replaced the A- and J-gates of Kane's concept by a linear addressing scheme using magnetic fields and a general SWAP implementation of electron–nuclear spin quantum computing. The implementation of universal quantum gates is then performed using only classical magnetic resonance pulses.

The concepts presented here will have to be verified experimentally step-by-step. An important first task is to measure the nuclear spin relaxation times and to try out the proposed SWAP operations between nuclear and electron spins. These experiments can be done on single molecules. The next steps involve the construction of a molecular spin quantum register bit-by-bit, which is already a quite formidable task necessitating the collaboration of several research groups. Finally, single spin read-out methods have to be developed in order to fully test the addressing scheme presented. If the basic ideas work out the scaling of the computer architecture should present only minor problems compared to the initial work outlined here.

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